

Fluctuation and Dissipation in Liquid Crystal Electroconvection

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In this experiment a steady state current is maintained through a liquid crystal thin film. When the applied voltage is increased through a threshold, a phase transition is observed into a convective state characterized by the chaotic motion of rolls. Above the threshold, an increase in power consumption is observed that is manifested by an increase in the mean conductivity. A sharp increase in the ratio of the power fluctuations to the mean power dissipated is observed above the transition. This ratio is compared to the predictions of the fluctuation theorem of Gallavotti and Cohen using an effective temperature associated with the rolls' chaotic motion.

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The fluctuation-dissipation theorem relates the spontaneous fluctuations in a system at temperature T with its response to an external driving force. An example might be a simple harmonic oscillator surrounded by air. The air molecules exponentially damp its periodic motion and also cause the oscillating mass to fluctuate about its equilibrium position [1].

The experiment described here is concerned, not with a system in thermal equilibrium but rather, one which is in a steady state: an electric potential V applied across a weakly conducting liquid crystal (LC), generates convective motion within it when V exceeds a critical value V_c [2]. The LC is in good contact with its surroundings, so that its temperature is fixed at the ambient value T , as it dissipates power P at a mean rate $\langle P \rangle$. Our interest centers on the dependence of $\langle P \rangle$ and its rms fluctuations σ_P as a function of V or, equivalently, the control parameter $\epsilon = ((V^2/V_c^2) - 1)$ [3].

Recently, Gallavotti and Cohen (GC) [4] have generalized the fluctuation-dissipation theorem (FDT) to include systems that are driven far from equilibrium, like the one studied here. While their work motivated the present experiment, we are unable to verify its central prediction concerning the probability density function $\pi(P)$. They showed that, under appropriate conditions, the system, assumed to be driven into a chaotic state, experiences such large fluctuations in P that sometimes P can become negative, i.e. the driven, dissipative system momentarily can send energy back into the power supply that generates the chaotic fluctuations. The theory makes a firm prediction about the ratio, $\pi(P)/\pi(-P)$.

The GC theory is formulated in terms of the entropy production rate $s_\tau = \dot{S}_\tau$, which is related to the average power gained by the system during a time period τ by $s_\tau = P_\tau/kT_{ss}$. Here the steady state temperature kT_{ss} is equal to the mean kinetic energy per particle. The authors show that if the chaotic motion in the system satisfies certain conditions, then

$$\pi(s_\tau)/\pi(-s_\tau) = \exp(\tau s_\tau).$$

The theorem, which GC call the fluctuation theorem (FT), has been generalized by Kurchan [5] to systems undergoing Langevin dynamics and by Lebowitz and Spohn to general Markov processes [6]. The FT reduces to the FDT in the limit of vanishing driving force [7].

For a macroscopic system like ours, it was not possible to achieve such large fluctuations in P that render the power negative. Thus we were barred from directly measuring the ratio $\pi(P)/\pi(-P)$ predicted by the GC theory. What we did find were unexpectedly large values of the dimensionless standard deviation $\sigma_P/\langle P \rangle$ when V exceeded V_c . Our results become consistent with predictions of the FT of GC, only if: (a) The temperature is taken to be the steady state temperature T_{ss} associated with the quasi-particles chaotic motion and not the ambient temperature T associated with the microscopic thermal motion and (b) the correlation length of the velocity fluctuations is taken to be much larger than the typical size of the rolls.

The liquid crystal used here is methoxy benzylidenebutyl aniline (MBBA), a nematic that undergoes a transition out of the amorphous state, when driven by an ac voltage, which will be written as $\mathcal{V}(t) = \sqrt{2}V \sin(2\pi ft)$. Typically f is in 100 Hz range, and the measurements reported here were made at exactly this frequency. The electrodes, across which this voltage is developed, are thin, transparent layers of indium-tin oxide, enabling their visual observation. When V exceeds the critical value $V_c = 6.0$ Volts in MBBA, the convective motion of the LC commences. The resulting convective motion is analogous to Rayleigh-Benard convection, but with the applied voltage V replacing the temperature difference across the sample [8].

Most studies of electroconvection in LC's center on the study of the patterns generated by the LC rather than the conductive behavior itself. Observations through a microscope reveal that above V_c , the convective rolls form a stationary pattern. At a very slightly higher voltage, the dislocations develop and, with a further voltage increase, commence to move. At even higher values of V/V_c , the

orientational fluctuations take on a chaotic or turbulent appearance [2] and the orientational domains fluctuate rapidly, though their motion is slow enough for the eye to follow. Thus this driven system, that appears to be in a chaotic steady state out of equilibrium, is a candidate for checking the applicability of the FT.

In the present experiment, the thicknesses d of the MBBA samples were 37.5 and 50 μm . These samples were square with the lateral dimensions L ranging from mm to cm. It was possible to make samples with L less than 1 mm, but their resistance was so high (hundreds of $\text{M}\Omega$) that measurements of the fluctuations in P could not be reliably made.

The experimental arrangement is extremely simple; the driving voltage $V(t)$, obtained from a signal generator, is applied across the sample, of resistance R , which is in series with a much smaller resistance r , which in our case was usually 100 $\text{K}\Omega$. The time-varying voltage v_r is pre-amplified and then fed into a lock-in amplifier tuned to the driving frequency f . The lock-in output is fed into dc amplifier (PAR 113) before it enters the A/D input of a computer, which records the detected signal. The current through the sample, $i = v_r/r$, and hence the power $P = iV$ fluctuates slowly compared to $1/f$, as we will see. The full spectrum of the fluctuations can be captured by setting the lock-in time constant at $\tau = 0.1$ s. [2].

Whereas this experiment is mainly concerned with fluctuations in P about its mean value, this latter quantity itself exhibits an interesting dependence on the control voltage V . Because $r \ll R$, one might expect that $\langle P \rangle = V^2/R$, so that a plot of $\sqrt{\langle P \rangle} \text{ vs } V$ would yield a straight line of slope $1/R$, and indeed this is so. But a more sensitive way of displaying the dependence of $\langle P \rangle$ on V is to plot the sample conductance $G \equiv \langle P \rangle/V^2$ as a function of V .

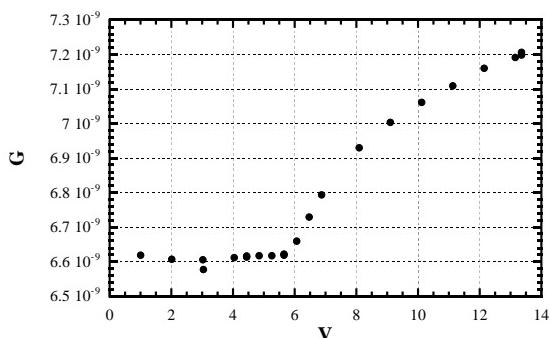


FIG. 1. Conductance $G = \langle P \rangle/V^2$, of MBBA sample in inverse Ohms. The control variable is the rms voltage, of frequency $f = 100$ Hz. The sample is a square of dimensions $L = 4$ mm and with $d = 50 \mu\text{m}$.

Figure 1 is a graph of this type. For $V < V_c = 6$ V, G is independent of V , and the sample behaves like a resistor, with $R = 1/G = 1/[6.6 \times 10^{-9}] = 150 \text{ M}\Omega$. This resistor-like behavior was verified by replacing the sam-

ple with a metal-oxide film resistor having $R = 100 \text{ M}\Omega$. One sees from this figure that the convective motion generates increased energy dissipation in the sample when V exceeds $V_c = 6$ V.

This appears to be the evidence that a phase transition to a chaotic state enhances the entropy generation rate s_τ (see above). The time interval τ in this experiment is the integration time of the lock-in, which is $\tau = 0.1$ s. Note that the vertical scale in Fig. 1 spans a very narrow range in G . It is not surprising then, that the transition at V_c was not detected in a similar measurement of the conductivity as a function of V in MBBA [2].

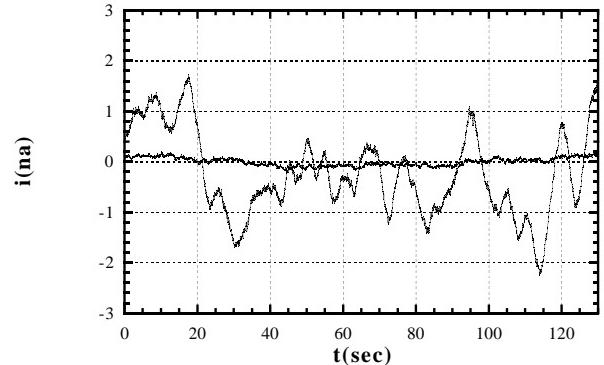


FIG. 2. Current through sample in nanoamps with its mean value subtracted out and a linear slow drift removed as well. The strongly fluctuating signal was recorded at $V = 1.67 V_c$. The weak signal is at $V = 4$ volts = $0.667 V_c$. The lateral dimensions of this square sample are $L=2.2$ mm, with $d=37.5 \mu\text{m}$.

Turning now to the *fluctuations* in P about its mean value, Fig. 2 shows a time trace of $i(t)$ at two applied voltages: $V = 4.0$ volts (weakly fluctuating signal) and $V = 10.0$ V (large, slow fluctuations). The sample thickness here is $d = 37.5 \mu\text{m}$, and its lateral dimensions are 2.3 mm and 2.15 mm. Over the two-minute interval of the measurements, the mean current drifts slightly, an effect probably caused by a slight drift in room temperature. The sense of the drift varies in sign for different measurements. To compensate for this drift, a term linear in i is subtracted out.

We have also measured the probability density function of the power dissipation P at various values of V . There was no systematic departure from a Gaussian form for this function $\pi(P)$; its kurtosis was quite close to the gaussian value of 3. The accuracy of these measurements is limited by the slow drift in the lock-in output mentioned above. It suffices then, to report the width σ_P of this function.

Figure 3 shows the non-dimensional standard deviation (STD) of the power fluctuations

$$\sigma_P / \langle P \rangle = \sqrt{\langle (P - \langle P \rangle)^2 \rangle / \langle P \rangle^2}$$

as a function of V . The sample is square with $L = 4$ mm and $d = 50 \mu\text{m}$.

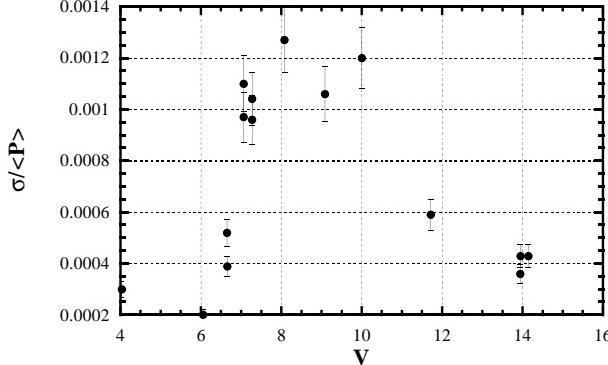


FIG. 3. Dimensionless standard deviation of the power fluctuations, $\sigma_P / \langle P \rangle$ vs V . For this sample $L = 4$ mm and $d = 50 \mu\text{m}$.

To interpret the power fluctuation data in Fig. 3, we assume that the noise that appears at $V < V_c$ is uncorrelated with the much slower fluctuations of interest above V_c and, being of small magnitude, we can neglect its contribution to $\sigma_P / \langle P \rangle$. We will also assume that above V_c the bulk of the power is dissipated as a result of the convective motion.

In the voltage interval where $\sigma_P / \langle P \rangle$ is large, the fluctuation time is significantly longer than outside that interval. This observation was difficult to quantify, even though it is apparent from looking at a record of $i(t)$. For example, the fluctuation rate appears larger than 10 Hz (the inverse of the lockin time constant) at $V = 4$ V, whereas it is smaller than 0.2 Hz at $V = 8$ V. At 14 V the fluctuation rate has increased to roughly 1 Hz.

The FC theorem of Gallavotti and Cohen concerns the rate of entropy production s_τ in thermostatted systems averaged over an observation time τ . The power dissipation can then be written as $P = s_\tau T_{ss}$, where T_{ss} is a temperature characterizing the kinetic energy of the fluctuations in the chaotic or turbulent steady state. Making certain assumptions about the time-reversal invariant chaotic motion that is generated by the driving force, Cohen and Gallavotti obtained the FT theorem as stated in the introduction. If we further assume that the power fluctuations about the mean are gaussian,

$$\pi_\tau(P) \propto \exp[-(P - \langle P \rangle)^2 / (2\sigma_P^2)]$$

as they appear to be, we find that

$$\frac{\pi_\tau(P)}{\pi_\tau(-P)} = \exp(2\langle P \rangle P / \sigma_P^2).$$

Comparing this to GC result

$$\frac{\pi_\tau(P)}{\pi_\tau(-P)} = \exp(\tau P / kT_{ss}),$$

we find that the dimensionless STD is given by

$$\sigma_P / \langle P \rangle = \sqrt{2k_B T_{ss} / \langle P \rangle \tau}. \quad (1)$$

To interpret our measurements in the convective region, $V > V_c = 6.0$ V, we will assume that the convective noise is uncorrelated with the noise that appears at, say, $V = 4$ volts, which is well below V_c , and we subtract it out in a definition of the *excess* noise $E(V)$:

$$E(V) = [\sigma_P(V) - \sigma_P(V = 4V)] / [\langle P \rangle - \langle P(V = 4V) \rangle]$$

To account for the measured maximum value of $\sigma_P / \langle P \rangle$ ($\simeq 10^{-3}$), we replace it with $E(V)$ and define a “convective temperature” T_{ss} by

$$k_B T_{ss} = (1/2) M v_{rms}^2, \quad (2)$$

where M is an appropriate mass of quasi-particles participating in the chaotic motion, and v_{rms} is the characteristic velocity of the local convective motion of the LC. We emphasize that T_{ss} differs from the molecular temperature, which is very close to the ambient temperature (~ 300 K), since the chaotic degrees of freedom are not in thermal equilibrium with the microscopic degrees of freedom.

By observing the trajectories of small particles convected by the motion of the LC [9,10], one finds that, in order of magnitude, $v_{rms} = v_0 \epsilon$, where $v_0 = 16 \mu\text{m/s}$. Our own admittedly crude measurements of the motion of dust particles give $v_{rms} \simeq 400 \mu\text{m/s}$ at $V = 9.6$ V for the 4 mm sample used in Fig. 3.

The diameter d of a roll is roughly the plate spacing, and in our samples their length was typically five times as large. For the sample of Fig. 3 this gives M in Eq. (2) $\simeq 10^{-9}$ Kg, since the specific gravity of MBBA is close to unity. Taking $v_{rms} \approx 100 \mu\text{m/s}$ one obtains $2k_B T_{ss} = 10^{-7}$ J. Using $\tau = 0.1$ s and the measured value of $\langle P \rangle$ at $V = 10$ V which is about 7×10^{-7} W, the rhs of Eq. (1) becomes approximately 10^{-5} , which is smaller by two orders of magnitude than the measured $\sigma_P / \langle P \rangle$. To accommodate for this difference, we argue below that the motion of the rolls may be correlated over scales much larger than the size of an individual roll.

The transition to the convective state is a continuous one as seen in Fig. 1; it resembles a continuous magnetic phase transition in a system in thermodynamic equilibrium or a supercritical Hopf bifurcation [11]. The “ordered phase” occurs for $V > V_c$. Recently it has been claimed [12] that a finite turbulent system is critical at all Reynolds numbers. It thus resembles a two-dimensional XY model which is characterized by a diverging correlation length throughout the ordered phase. This is because all length scales between the size of the system, where the driving occurs, to the microscopic scale, where dissipation occurs, are connected by the cascading process. Thus, we would expect the system to exhibit a large correlation length (of the order of the system size) above

the onset of convection. This may provide a possible explanation for the large magnitude of the fluctuations. If the rolls exhibit a collective correlated motion, then the relevant mass could be as large as the total mass of the liquid crystal. This will increase our estimate of Ω sufficiently to bring the measurement and the calculation of $E(V)$ into approximate agreement. The correlation length associated with the collective motion of the rolls should be distinguished from the much smaller length that is associated with the director fluctuations. This state of the LC in the present experiment is of course not turbulent, because the Reynolds number Re associated with the roll size and v_{rms} is much less than unity, but the motion is nevertheless chaotic. Hence the present experiment is very different from that discussed in Ref. [12], where the Reynolds number was large.

To establish whether the numerator in Eq. (1) is indeed proportional to the mass M of entire sample, we measured $\sigma_P/\langle P \rangle$ and $\langle P \rangle$ in two samples having different lateral dimensions, $L = 2$ mm and 11 mm. Both measurements were made at the same voltage $V = 10.1$ V, so that v_{rms} presumably remains the same. According to the above model, $\sigma_P/\langle P \rangle \propto L/\sqrt{\langle P \rangle}$. The ratio $R = (\sigma_P/\langle P \rangle)_{2mm}/(\sigma_P/\langle P \rangle)_{11mm} = 1/2$. This is to be compared with the measured ratio, $R = 2$. To resolve this inconsistency, we must assume that the correlation length of the power fluctuations saturates at a scale of the order of a mm. With this assumption, the measured and calculated values of R come into approximate agreement.

In summary, we have measured the steady-state power dissipation $\langle P \rangle$ in a fluid system (thin layer of liquid crystal) in the vicinity of a phase transition from the quiescent to the convective state, where the fluid motion of the MBBA liquid crystal presumably becomes chaotic. To maintain this chaotic motion at voltages exceeding the critical value, the system extracts energy from the power supply at an increasing rate, implying an increase in its mean conductivity $G(V)$.

The dimensionless width of the fluctuations, $\sigma_P/\langle P \rangle$, is much larger than that associated with a resistor at ambient temperature T but goes through a maximum near $V \simeq 2V_c$. The magnitude of $\sigma_P/\langle P \rangle$ near its maximum value can be approximately understood in terms of the fluctuation-dissipation theorem and the fluctuation theorem of Gallavotti and Cohen, provided that one inserts into Eq. (1) a temperature kT_{ss} that is identified with the kinetic energy of the quasi-particles excited by the flow. If the mass of these excitations is taken to be that of the convective rolls in the system, the dimensionless width of the power fluctuations is somewhat smaller than the measured value. An even better agreement is achieved if one assumes that the motion of the rolls is correlated with a correlation length much larger than the size of the rolls observed with a microscope. It thus appears that the mass of the excitations in a measurement of the power fluctuations is an appreciable fraction of the mass of the liquid crystal.

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Note added: After this work was completed we received a preprint from J. T. Gleeson et al. [13] showing a transition similar to that depicted in Fig 1.

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